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FINAL REPORT

ACOUSTIC CAVITATION STUDIES

September 1981

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Lawrence A. Crum
Department of Physics and Astronomy
University of Mississippi
Oxford, Mississippi 38677

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Introduction:

This final report constitutes a review of the work accomplished under contract N00014-79-C-0404 of the Office of Naval Research. The primary thrust of this study was toward a more complete understanding of general aspects of acoustic cavitation. The specific aspects of this study are discussed below.

Studies in Cavitation Inception

The effect of long-chain polymer additives on the cavitation threshold was investigated to determine if they reduced the acoustic cavitation threshold in a similar manner to the observed^{1,2} reduction in the cavitation index in hydrodynamic cavitation. Measurements were made of the acoustic cavitation threshold as a function of polymer concentration for additives such as guar gum and polyethelene oxide. The measurements were also made as a function of dissolved gas concentration, surface tension and viscosity. It was determined that there was a significant increase in the acoustic cavitation threshold for increased concentrations of the polymer additives (measurable effects could be obtained for concentrations as low as a few parts per million). One would normally expect that an additive that reduces surface tension to decrease the pressure required to cause a cavity to grow and thus these additives, at first thought, should reduce the threshold. However, even in the hydrodynamic case, the threshold was increased. In both of the hydrodynamic cases considered, the explanation for the increased threshold was given "in terms of changed fluid dynamics rather than changed physical properties of the fluid." They based this explanation on what this author feels was an incorrect application of the general equations of cavitation nucleation.

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They also were unable to give a quantitative explanation of the threshold change.

We have analyzed our data of the acoustic cavitation threshold under the assumption that nuclei are produced from gas pockets trapped in particulate matter^{3,4} and have been able to obtain excellent agreement between theory and the threshold data. Some preliminary results are given in a review article on microbubble nucleation and and stabilization⁵ and it is planned to submit the full results for publication in the immediate future.

Studies in Nonlinear Bubble Pulsations

We have developed an experimental apparatus to measure properties of air bubbles that have been acoustically levitated (or antilevitated) in a standing wave system. We have used this apparatus before for rectified diffusion studies⁶. Using this system it was discovered that slight changes in the properties of the bubble pulsation could be detected as a shift in the equilibrium position of the levitated bubble. For example, for the same acoustic pressure amplitude, bubble radius, driving frequency, and host fluid, an air bubble filled with air would take a slightly different position than a bubble filled with a monatomic gas, say, such as argon. The pulsation amplitude is dependent upon the thermodynamic properties of the gas within the bubble and consequently we were able to measure this behavior. In particular, we were able to obtain values of the polytropic exponent of the gas that compared quite favorably with calculations. Another, even more interesting, observation was that the air bubble would be displaced slightly when it was driven nonlinearly in one of its harmonic frequencies. These nonlinear harmonic oscillations have been predicted by the analytical approach of Prosperetti⁷

and by the numerical approach of Lauterborn⁸. We have been able to see both the $n = 2$ and the $n = 3$ harmonics for air bubbles in a variety of liquids. Comparison with the analytical predictions of Prosperetti⁷ show some comforting agreements but also some really intriguing disagreements. In particular, there is an apparent shift in the resonance frequency of the air bubble as the amount of vapor in the interior of the bubble is increased (we have done this by raising the temperature or by using a highly volatile host fluid). There are some predictions by Prosperetti that the vapor will have an effect but just how the effect applies here has not been determined. We have opened a collaboration with Dr. Prosperetti, University of Milano, Italy, and have obtained a NATO travel grant to visit each other's laboratory this coming year. Although funds are no longer available to support this research from the ONR, we are currently seeking other funds to follow up on these very interesting and important discoveries. A preliminary report on this research was presented at a meeting of the Acoustical Society of America, (see publication A-1 in Appendix I) and we expect to generate a report of considerable length on this topic in the immediate future.

Studies in Rectified Diffusion

Whenever a sound field is present in a liquid, there will probably be some degree of rectified diffusion. Almost all liquids have present in them in different quantities stabilized pockets of air that will pulsate to some degree due to the presence of acoustic pressure waves. Associated with these pulsations will be rectified diffusion. We have examined this process because it is especially relevant to the general aspects of cavitation and because we have discovered that it can play an important role in the effect of ultrasound on living tissue. The

literature is full of the effects of moderately high intensity sound on biological systems and in many cases it appears that the indirect culprit is an air bubble. In our recent review paper⁵, we were able to show that several different biological effects that were observed during or after irradiation by ultrasound could be the result of air bubble interactions, and particularly those associated with bubbles that have grown to resonance size via rectified diffusion. Recently we have shown⁹ that air bubbles that have appeared in guinea pig legs during insonification at intensities as low as 80mW/cm^2 could be the result of rectified diffusion. We presently are preparing a comprehensive treatment of the role of rectified diffusion in biological systems and hope to have it submitted for publication before the end of the calendar year. A paper describing a generalization of the rectified diffusion equations¹⁰ has been submitted to the Journal of the Acoustical Society of America for publication.

Summary:

This contract resulted in 8 external communications (three more are being planned in the immediate future) and with the essential completion of two MS degrees and about half of one Ph.D. In addition, six other students were partially trained in this program.

Financial Report:

The duration of the first contract period was from 15 May 1979 through 14 May 1980 funded in the amount of \$29,549, and renewed, with a no funds extension through July 15, 1981 in the amount of \$37,930. The funds were totally expended essentially in accordance with the proposed budgets.

Personnel Report:

The names of students associated with this contract, their employment period, their current status, and degrees attained are listed below:

1. Bruce Hellman, May 1979 - September 1980. Mr. Hellman was a Ph.D. candidate who made some contributions to the theories of rectified diffusion and bubble pulsation. After failing his Ph.D. comprehensive exam in July, 1980 he chose to leave the University. (No degree obtained)
2. Emilio Ramos, April 1980 - July 1981. Mr. Ramos was an M.S. candidate who acquired much of the data on cavitation inception. He has subsequently transferred to another university. (No degree obtained)
3. James Brosey, April 1980 - present. Mr. Brosey is an M.S. candidate who completed the work of Mr. Ramos, and is presently writing his thesis. (M.S. expected, December, 1981)
4. David Young, April 1980 - present. Mr. Young is an M.S. candidate who acquired the data on bubble pulsation amplitude. He finished his course work, has taken a job and plans to return next summer to write his thesis. (M.S. expected, August, 1982)

5. Gary Hansen, April 1980- present. Mr. Hansen is a Ph.D. candidate who has done much of the theoretical work associated with this contract. He will take his Ph.D. comprehensive exam this fall and has already made considerable progress toward a doctoral dissertation. (Ph.D. expected, August, 1984)

In addition the following undergraduate students were hired at various times to assist in some laboratory tasks and to gain research familiarization: (a) Ron Roy, (b) Kerry Commander, (c) Tim Farris and (d) Mike Lipski.

List of References

1. A.T. Ellis, J.G. Waugh and R.Y. Ting, "Cavitation Suppression and Stress Effects in High-speed Flows of Water with Dilute Macromolecule Additives." J. Basic Engr., Trans. ASME 92, 459-466 (1970).
2. J.W. Hoyt, "Effect of Polymer Additives on Jet Cavitation," J. Fluids Engr. 98, 106-112 (1976).
3. L.A. Crum, "Tensile Strength of Water," Nature, 278, 148-149 (1979).
4. L.A. Crum, "Acoustic Cavitation Thresholds in Water," in Cavitation and Inhomogeneities in Underwater Acoustics, ed. by W. Lauterborn, (Springer-Verlag: New York) 1980.
5. L.A. Crum, "Nucleation and Stabilization of Microbubbles in Liquids," App. Sci. Res. (in press).
6. L.A. Crum, "Measurements of the Growth of Air Bubbles by Rectified Diffusion," J. Acoust. Soc. Amer. 68, 203-211 (1980).
7. A. Prosperetti, "Nonlinear Oscillations of Bubbles in Liquids," J. Acoust. Soc. Amer. 56, 878-885 (1974).
8. W. Lauterborn, "Numerical Investigation of Nonlinear Oscillations of Gas Bubbles in Liquids," J. Acoust. Soc. Amer. 59, 283-293 (1976).
9. L.A. Crum and G.M. Hansen, "Growth of Air Bubbles in Tissue by Rectified Diffusion," Phys. Med. and Biol. (submitted for publication).

APPENDIX I

LIST OF EXTERNAL COMMUNICATIONS
ORIGINATED UNDER CONTRACT

A. Papers presented:

1. "The Pulsation Amplitude of Gas Bubbles in a Stationary Sound Field," with G.E. Herring, Jr. and D.A. Young, presented at the 100th meeting of the Acoustical Society of America, Los Angeles, November, 1980.
2. "Rectified Diffusion at Megahertz Frequencies," with G.M. Hansen, presented at the 101st meeting of the Acoustical Society of America, Ottawa, May, 1981.
3. "Nucleation and Stabilization of Microbubbles in Liquids," an invited paper presented at the International Union of Theoretical and Applied Mechanics (IUTAM) symposium on the physics of bubbles in liquids, Pasadena, June, 1981.

B. Technical Reports submitted:

1. Air Bubble Growth by Rectified Diffusion, Technical Report No. 802 under Office of Naval Research Contract N00014-79-C-0404, University of Mississippi, April, 1980.

C. Articles submitted for publication:

1. "Measurements of the Growth of Air Bubbles by Rectified Diffusion," J. Acoust. Soc. Amer. 68, 203-211 (1980).
2. "Nucleation and Stabilization of Microbubbles in Liquids," App. Sci. Research, accepted for publication.
3. "Growth of Air Bubbles in Tissue by Rectified Diffusion," submitted to Physics in Medicine and Biology.
4. "Generalized Equations of Rectified Diffusion," submitted to Journal of the Acoustical Society of America.

APPENDIX II

ABSTRACTS OF EXTERNAL COMMUNICATIONS
ORIGINATED DURING CONTRACT PERIOD

The pulsation amplitude of gas bubbles in a stationary sound field. L.A. Crum,
C.E. Herring, Jr. and D.A. Young, Department of Physics and Astronomy
University of Mississippi, Oxford, MS 38677.

It has been known for some time that individual gas bubbles can be trapped near the pressure antinode of a stationary sound field provided they are driven below their resonance frequency. The acoustic force that balances the buoyant force is proportional to the pulsation amplitude of the bubble. This pulsation amplitude can be ascertained indirectly through measurements of the acoustic pressure amplitude, the bubble radius, and the position of the bubble in the sound field. Further, the theoretical pulsation amplitude can be obtained by a solution of the well-known "Rayleigh-Plesset-Noltingk-Neppiras-Poritsky equation" that has been used extensively in cavitation research. We present measurements of the pulsation amplitude of bubbles filled with various gases and in a variety of liquids at a frequency of 20 kHz. We also have obtained analytical solutions of the RPNNP equation by an expansion technique. Comparison of theory and experiment show good agreement for relatively small radii but substantial disagreement as the bubble radius approaches the resonance radius. It is thought that the source of the disagreement is the presence of surface waves on the bubble. (Work supported by the Office of Naval Research)

Presented at the 100th meeting of the Acoustical Society of America, Los Angeles, November, 1980.

Rectified Diffusion at Megahertz Frequencies. Lawrence A. Crum and Gary M. Hansen, Department of Physics, University of Mississippi, Oxford, MS 38677

Rectified diffusion studies have in the past been generally confined to kilohertz frequency ranges where experimental confirmation of theoretical predictions can be made. However, since most of the ultrasonic devices used in medicine operate in the megahertz frequency range, it is important to extend these studies of the growth of bubbles by ultrasound to the higher range. We have modified and extended the equations associated with rectified diffusion to apply at megahertz frequencies, and have numerically solved these equations for a variety of conditions such as dissolved gas concentration, distribution of nuclei, and the frequency and intensity of the ultrasound. We have also obtained solutions for both the continuous and pulsed modes of operation. Our results indicate that bubbles can be made to grow at typical levels of exposure used in medical applications. Furthermore, at higher intensities, such as those used in studies on bean roots by Morris and Coakley (to be published in *Ultrasound in Med. and Biol.*), the times required for growth to resonance size are on the same order of magnitude as the periods associated with acoustic emissions from the root during insonification. (Work partially supported by the Office of Naval Research and the National Science Foundation.)

Presented at the 101st meeting of the Acoustical Society of America, Ottawa, May, 1981.

Nucleation and Stabilization of Microbubbles in Liquids

Lawrence A. Crum, Department of Physics and Astronomy, University of Mississippi, Oxford, Mississippi 38677

The measured tensile strength of normal liquids is commonly found to be significantly less than that predicted on the basis of intramolecular forces. There have been some measured values of liquid tensile stress that are comparably to that predicted on the basis of homogeneous nucleation theory, but only for liquids that have been contained in extremely minute samples (Apfel, R. E., *Nature* 233, 119-121 (1971).). It is reasoned that these samples are so small that there is a small probability that they will contain an impurity that can provide a preferential site for liquid rupture. This paper reviews the various models that have been proposed for the introduction of nucleation sites into the liquid and the subsequent stabilization of these nuclei from dissolution. Here it is assumed that the nuclei are small concentrations of gas contained within a small but well defined volume within the liquid. The nuclei are often termed microbubbles. An effort will be made to critically examine these models on the basis of the available experimental evidence and to show how some models may be rejected, others accepted, and still others that can not be grouped into either category. Finally, one particular model that is favored by the author will be examined in some detail and results will be presented that will lend considerable support toward its acceptance as a viable mechanism for microbubble nucleation and stabilization. (Work supported by the Office of Naval Research and the National Science Foundation.)

An invited paper presented at the International Union of Theoretical and Applied Mechanics (IUTAM) symposium on the physics of bubbles in liquids, Pasadena, June, 1981.

Air Bubble Growth by Rectified Diffusion

Lawrence A. Crum, Department of Physics and Astronomy, University of Mississippi, Oxford, Mississippi 38677

Measurements are reported of the growth of air bubbles in water by rectified diffusion at 22.1 kHz. Values of the threshold acoustic pressure amplitude were obtained as a function of bubble radius, liquid surface tension and gas concentration. Measurements of the rate of growth of bubbles by rectified diffusion were also obtained as a function of acoustic pressure amplitude for a range of different values of the liquid-vapor surface tension. It was determined that although both the threshold and the growth rate were in agreement with theory for normal values of the surface tension of water, the addition of a surfactant caused the observed thresholds and growth rates to deviate from the predicted values. Surface wave activity that could increase the diffusion rate by acoustic streaming was not detected at low radii and is not thought to be the principal mechanism for the increased diffusion. Some possible explanations are given for the effect.

ONR Technical Report #802 under contract N00014-79-C-0404.

Nucleation and Stabilization of Microbubbles in Liquids

Lawrence A. Crum, Department of Physics and Astronomy, University of Mississippi, Oxford, Mississippi 38677

An important aspect of the processes of cavitation and boiling is the concept of a nucleus that acts as a preferential site for the inception of these events. It is commonly thought that except for rare instances or specially controlled experiments, all cavitation and boiling sites originate at the location of such a nucleus. In order to study these important phenomena, then, it is imperative that as much as possible be known about nucleation in cavitation and boiling. It is generally accepted that free air bubbles normally do not act as nucleation sites because they are inherently unstable to dissolution due to surface tension. Thus, the study of nucleation is necessarily associated with mechanisms for stabilizing microbubbles or pockets of gas within the liquid. In this paper, various stabilization models that have been proposed are reviewed as well as the experimental evidence that supports the specific models. One particular model, the crevice model, is examined in some detail, and its predictions are used to explain several different measurements of boiling and cavitation inception. Finally, some evidence that has recently become available concerning the damaging aspects of high intensity ultrasound is examined. Many aspects of this evidence point to the existence of cavitation as the damage mechanism. Also given in this paper are preliminary explanations of these effects due to the growth of microbubbles or cavitation nuclei by rectified diffusion.

Accepted for publication in Applied Scientific Research.

Measurements of the Growth of Air Bubbles by Rectified Diffusion

Lawrence A. Crum, Department of Physics and Astronomy, University of Mississippi, Oxford, Mississippi 38677

Measurements are reported of the growth of air bubbles in water by rectified diffusion at 22.1 kHz. Values of the threshold acoustic pressure amplitude were obtained as a function of bubble radius, liquid surface tension and gas concentration. Measurements of the rate of growth of bubbles by rectified diffusion were also obtained as a function of acoustic pressure amplitude for a range of different values of the liquid-vapor surface tension. It was determined that although both the threshold and the growth rate were in agreement with theory for normal values of the surface tension of water, the addition of a surfactant caused the observed thresholds and growth rates to deviate from the predicted values. Surface wave activity that could increase the diffusion rate by acoustic streaming was not detected at low radii and is not thought to be the principal mechanism for the increased diffusion. Some possible explanations are given for the effect.

Published in Journal of the Acoustical Society of America, 68, 203-211 (1980).

Growth of Air Bubbles in Tissue by Rectified Diffusion

Lawrence A. Crum, Department of Physics and Astronomy, University of Mississippi, Oxford, Mississippi 38677

A recent report [ter Haar and Daniels, Phys. Med. and Biol. (in press)] has presented evidence for ultrasonically induced cavitation in vivo. In this experiment, guinea pig legs were irradiated with continuous ultrasound at spatial average intensities varying from 80 mW/cm² to 680 mW/cm². In addition, the leg was scanned with a pulse-echo ultrasonic imaging system in order to detect both moving and stationary cavitation bubbles down to 10 μ m in diameter. The results of their experiment were that air bubbles of detectable size (that is, larger than 10 μ m in diameter) were generated in the guinea pig tissues at intensities as low as 80 mW/cm² and that progressively more bubbles were detected as the intensity was increased to 680 mW/cm². This paper will demonstrate that the observed production of stable cavitation bubbles in tissue, at the frequency and intensities used by ter Haar and Daniels, can be predicted on the basis of a presently available theory concerning the growth of air bubbles by rectified diffusion.

Submitted for publication to Physics in Medicine and Biology.

Generalized Equations of Rectified Diffusion

Lawrence A. Crum and Gary M. Hansen, Department of Physics and Astronomy,
University of Mississippi, Oxford, Mississippi 38677

There are published articles by several authors that introduce equations that predict the growth rate and the threshold for growth of air bubbles by rectified diffusion. Most of these equations are restricted to special circumstances, and neglect such things as surface tension, the thermodynamic behavior of the gas in the interior of the bubble, and the damping of the bubble pulsations. We have generalized the equations of rectified diffusion so that they should now apply over a broad range of applicable parameters. We have also shown that the equations given in previous publications are special cases of our more generalized case. Finally, we have graphically displayed the predictions of the various equations, including our more complete, but also more complicated, result, in order to show where the simpler equations can be applied with small error.

Recently submitted to the Journal of the Acoustical Society of America.

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